

JP62278770

Publication Title:

RECOVERING METHOD OF CO₂ AND H₂ FROM CONVERTER GAS

Abstract:

PURPOSE: To increase utilization by treating converter gas with a pressure swing adsorber (PSA), treating again with a PSA installed in series, and supplying the obtained H₂ to a fuel cell to generate electric power and recovering exhaust heat as steam.

CONSTITUTION: LD converter gas is modified to H₂, and treated with a pressure swing adsorber (PSA) 3 through a absorbin tower 1, and offgas RH₂ which contains other gasses in addition to H₂ is stored in a holder 4. The offgas RH₂ is treated with a PSA 6 installed in series to obtain H₂. The H₂ obtained is supplied to a negative electrode 13 of a fuel cell 15 and reacted with oxygen in a positive electrode 14 to generate electric power. Exhaust gasses from the negative electrode 13 and the positive electrode 14 are supplied to a combustor 18 to completely burn and exhaust heat is recovered in a boiler 19. Therefore, electric power, steam, and fuel gas can be obtained from offgas from purification equipemnt of converter gas, and utilization is increased.

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⑫ 公開特許公報 (A) 昭62-278770

⑮ Int.Cl.

H 01 M 8/06
C 21 C 5/38
// C 10 K 1/12
1/20

識別記号

府内整理番号

R-7623-5H
Z-6813-4K
6683-4H
6683-4H

⑯ 公開 昭和62年(1987)12月3日

審査請求 未請求 発明の数 1 (全4頁)

⑰ 発明の名称 転炉ガスからのCO₂, H₂回収方法

⑱ 特願 昭61-120218

⑲ 出願 昭61(1986)5月27日

⑳ 発明者 藤井 史朗 東京都千代田区丸の内1丁目1番2号 日本钢管株式会社
内

㉑ 出願人 日本钢管株式会社 東京都千代田区丸の内1丁目1番2号

㉒ 代理人 弁理士 佐藤 正年 外2名

明細書

1. 発明の名称

転炉ガスからのCO₂, H₂回収方法

2. 特許請求の範囲

転炉ガスを変成し、次いで該変成ガス中のCO₂を吸収塔にて吸収し、該吸収液よりCO₂を取得し、次いで、該吸収塔オフガスを昇圧し、昇圧ガスをPSA (圧力式吸着装置) にて処理することにより、CO₂及びH₂を回収する方法において、該PSAオフガスを、更に直列に設けたPSAにて処理した後に燃料電池に供給し、電力を取得し、該燃料電池のオフガスを完全燃焼せしめ、燃焼熱をステームに変換利用することを特徴とする転炉ガスからのCO₂, H₂回収方法。

3. 発明の詳細な説明

(産業上の利用分野)

本発明は、LD転炉等から製鋼時発生する転炉ガスからのCO₂, H₂を回収し、熱及び電気エネルギーを取得する方法に関するものである。

(従来の技術)

近時、製鋼用LD転炉等においては、操業時発生する、CO₂, CO₂及びH₂を含有する転炉排ガスを積極的に回収し有効利用を図っている。

これら転炉ガス成分は、容積基準(以下同じ)にて、CO₂10~15%, H₂1%, CO 65~75%, N₂10%, O₂1%である。

この転炉ガスは、それ自体は燃料ガスとして使用し得るものであるが、更にこのガスを原料ガスとして、これに水蒸気を添加しシフト反応を生ぜしめ、二酸化炭素及び水蒸気とし、付加価値を向上せしめる回収方法が一部に行われている。

第2図は、LD転炉ガスから、CO₂及びH₂を回収するための従来法の工程図である。

図において、前記組成のLD転炉ガスを原料として、該ガスを9~20kg/m³に昇圧し、これに水蒸気を添加しシフト反応を起せしめる変成工程を経て、ガス成分を変成せしめる。(なお以上については図示なし。)

次いで吸収塔1に変成ガスを導入し、吸収液としての炭酸カリ液(K₂CO₃)中に、変成ガス中

のCO₂を吸収せしめ炭酸水素加里 (K₂CO₃) とし、これを水蒸気にてストリッピングし、吸収液はK₂CO₃とし、一方ストリッピングされたCO₂ガスを製品とする。

CO₂を放出し、H₂リッチの吸収塔1のオフガスは、PSAコンプレッサー2にて9~20kg/cm²から9~25kg/cm²に昇圧し、PSA (Pressure Swing Adsorption圧力式吸着装置) 3にて処理する。PSAにては、水素以外の気体を、圧力差を利用して吸着し、製品高純度水素 (PH₂) とオフガス (RH₂) とに分離し、オフガスはRH₂ホールダー4に貯留し、ブローウェル5にて、他のプロセスの燃料用として送気している。

この場合のオフガスの成分例は、CO₂12%, H₂58%, CO 3.5%, N₂26%, O₂0.5%である。

〔発明が解決しようとする問題点〕

説上の如く、転炉ガスからCO₂, H₂を回収する従来の回収方法におけるオフガスは、H₂を約60%程度含有しているにも拘らず、單に燃料用としてのみ利用されているに過ぎない。

廃ガスを昇圧し、昇圧ガスをPSAにて処理することにより、H₂を分離回収する方法において、該PSAオフガスを、更に直列に設けたPSAにて処理した後のガス (CO成分が低くH₂成分の濃度が高いガス) を燃料電池に供給し、電力を取得し該燃料電池のオフガスを完全燃焼せしめ、燃焼熱をスチームに変換利用することを特徴とする転炉ガスからのCO₂, H₂の回収方法である。

〔作用〕

本発明の転炉ガスからのCO₂, H₂回収方法においては、水素を約60%含有するPSAオフガスを更に直列に設けたPSAにて処理し、CO成分の少ない、濃度の高いH₂を収率良く回収することができたので、燃料電池の燃料ガスとして供給を可能とし得て、

更に、燃料電池のオフガスを完全燃焼せしめその排熱をスチームとして回収でき、電力、スチーム、燃料ガスなどの多様エネルギーを一挙にかつコスト的に有利に回収することを可能とするものである。

製鉄所周辺の病院あるいは事務所等においては、スチーム、電力と共に給湯、冷暖房設備等が必要である。現状においては、前述の如く、転炉ガスからの回収CO₂, H₂等の有力なエネルギー源があるに拘らず、買電々力にて前述の様な福利、管理設備の需要をみたし、また給湯、冷暖房には重油等を使用しているケースが多い。

本発明は、転炉ガスからのCO₂, H₂製造設備の比較的カロリーが低い (500~1700KCal / Nm³) が、水素濃度の高いオフガスを効率的に回収し、かつその保有するエネルギーを効率的に利用する方法を提供することを目的とするものである。

〔問題点を解決するための手段〕

本発明者等は、従来法における水素約60%を含有しているPSAオフガスに着目し、このオフガスの有効利用を研究し本発明に至ったものである。即ち本発明は、

転炉ガスを変成し、次いで、変成ガス中のCO₂成分を吸収塔にてK₂CO₃液中に吸収し、該吸収液より製品CO₂を取得し、次いで該吸収

次に実施例について述べる。

〔実施例〕

第1図は、本発明の転炉ガスからのCO₂, H₂回収方法の工程説明図である。

第1図において、1~4は前述の第2図と同一なので説明を省略する。

6は直列に設けたPSA、7は真空ブローウェル、8はブローウェル、9はバルブ、10は熱交換器、11は燃焼用空気ブローウェル、12は燃料電池、13は水素極室 (負極)、14は酸素極室 (正極)、15は電解液、16はポンプ、17は予熱器、18は燃焼器、19はボイラー、20は煙突である。

第1図に基づいて、本発明方法について述べる。

LD転炉ガス (H₂: 0.7%, CO: 69.8%, CO₂: 14.7%, N₂: 14.3%) 1.298Nm³/hを原料ガスとして、このガスを9~11kg/cm²にコンプレッサーにて昇圧し、次いで、このガスを水蒸気を用いシフト反応により水素に変成し次いで吸収塔1にて、K₂CO₃液によりガス中のCO₂を

吸収し、吸収液を水蒸気にてストリッピングし、製品CO₂を得る。次いで吸収塔1のオフガスをPSAコンプレッサー2にて16kg/cm²に昇圧しPSA3にて処理する。

PSA3にては、塔内の吸着剤にH₂以外のガスを吸着せしめ、製品PH₂と、H₂以外のガスをも含むオフガスRH₂とに分離する。

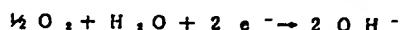
オフガスRH₂は一旦ホルダー4に貯留し、バルブ9を制御することにより、適宜プローワー8(2000mm水柱)にてオフガスを次のPSA6にて処理する。

PSA6からの処理水蒸ガス(553N m³/H)は、PSA6にて処理したためにH₂は89.5%, COは1.1%, CO₂は0.4%, N₂9%を含有し、燃料電池に供給しうるものである。

次に燃料電池から電力及び蒸気を回収する実施例について述べる。

上記PSA6からの処理水蒸ガスは真空プローワー7にて吸引し、熱交換器10にてオフガス温度を、燃料電池12のオフガスの燃焼排熱を回収

放し水酸イオンと酸素になり、この酸素は再び電極反応に利用される。従って正極の反応は



この正極14で生成した水酸イオンは、電解液15中を移動して負極13に達し、全体の回路を形成し電力740kWを取得した。

斯かる燃料電池の負極13及び正極14からの夫々190°Cオフガスは燃焼器18に導入され、完全燃焼し、430°Cの燃焼排ガスは、ボイラーレ19にてその発熱を回収し6kg/cm²の飽和蒸気0.7T/Hを回収した。

なお負極13室内には純水の予熱器17を設け予熱後ボイラーレ19に供給する。

ボイラーレ19のオフガスは熱交換器10にて前述の如く燃料電池への供給ガス及び空気を予熱後煙突20より排煙する。

以上の結果、転炉ガスの評価が従来法に比して1.86倍の価値となつた。

(発明の効果)

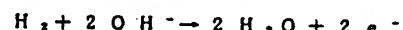
本発明の転炉ガスからのCO₂, H₂回収方法に

後の廃ガスと熱交換し90°Cに昇温し、燃料電池12の水素極(負極)13に導入する。

一方空気2000N m³/Hをプローワー11にて熱交換器10に供給し、120°Cに昇温し該空気を燃料電池12の酸素極14に導入する。

燃料電池12は、リン酸水溶液15を電解液とし、これを挿んで、白金、ルテニウムからなる2つの電極から構成されている。

負極の水素電極13では、導入された水素ガスが、電極内の細孔を通って反応帯域近くに達し、電極に添加されている触媒に吸着され、活性な水素原子となる。この水素原子は次式のように、電解液中の水酸イオンと反応して水となり、その際2個の電子を電極に送る。



正極の酸素極14では、触媒の存在の上で、電極から2個の電子をうけとり、外部から供給された空気中の酸素分子が、電解液からの水と反応して次式のように、過酸化水素イオンと水酸イオンを生成し、過酸化水素イオンは触媒と接して分

よれば次の如きの効果を及ぼすものである。

- ① 転炉ガスの精製設備からのオフガスより電力、蒸気、燃料ガス等を一挙に取得でき、転炉ガスの高付加価値化が可能である。
- ② 電力、蒸気、燃料等の多面的需要に対応できるので、製鉄所周辺の設備合理化が計られる。

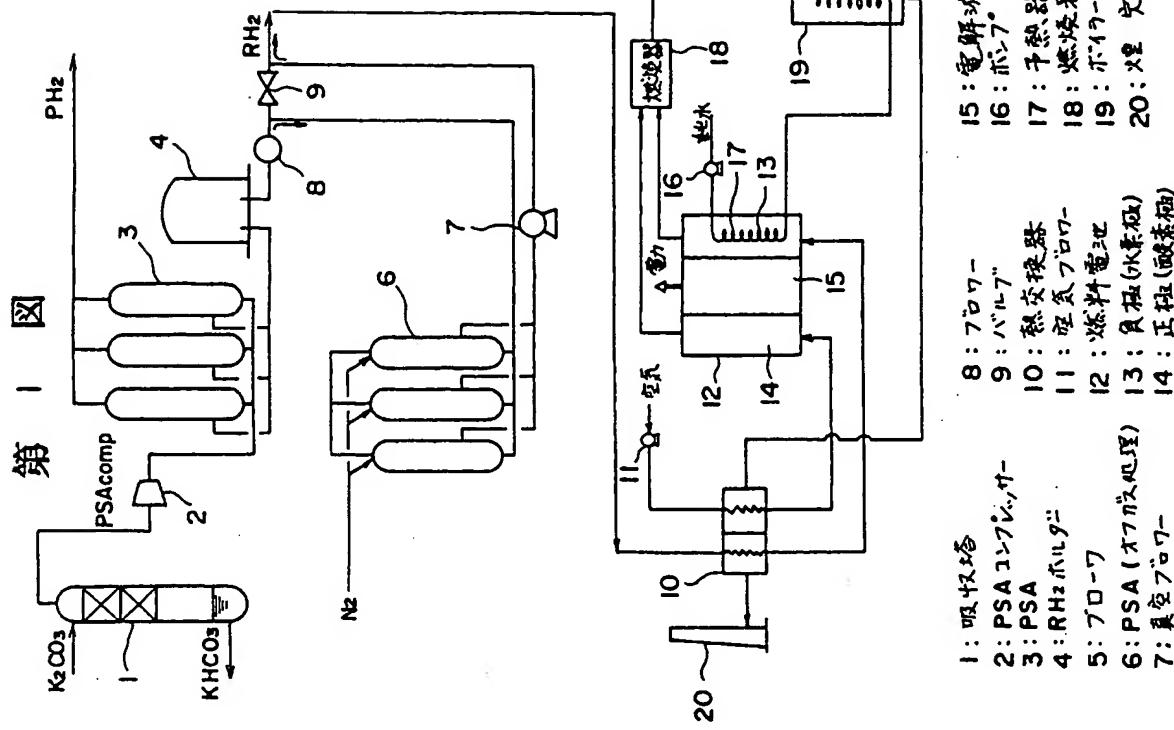
4. 図面の簡単な説明

第1図は本発明方法の工程説明図、第2図は従来法における工程説明図である。

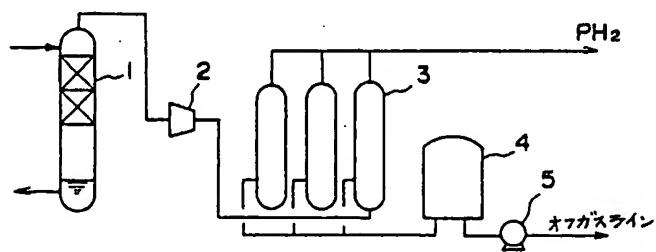
図において1:吸収塔、2:PSAコンプレッサー、3:PSA、4:RH₂ホルダー、6:PSA(オフガス処理)、7:真空プローワー、8:プローワー、9:バルブ、10:熱交換器、11:空気プローワー、12:燃料電池、13:負極(水素極)、14:正極(酸素極)、15:電解液、17:予熱器、18:燃焼器、19:ボイラーレである。

なお、各図中同一符号は同一または相当部分を示す。

代理人弁理士佐藤正年



第2図



Japanese → English Laid-open Patent Application No. Sho 62-278770
Klarquist, Sparkman, LLP Ref. No. 6454-58227

19. Japan Patent Office (JP)

12. Laid-open Patent Application Gazette (A)

11. Laid-open Patent Application No. Sho 62-278770

43. Disclosure Date: December 3, 1987

51. Int. Cl.⁴ ID Code JPO Control No.

 H 01 M 8/06 R-7623-5H

 C 21 C 5/38 Z-6813-4K

 // C 10 K 1/12 6683-4H

 1/20 6683-4H

Number of Inventions: 1

Examination: Not Requested Yet

Total 4 pages

54. Title of Invention: Method of Recovering CO₂ and H₂ from
 Converter Gas

21. Application No. Sho 61-120218

22. Application Date: May 27, 1986

72. Inventor: Shirou Fujii, Nippon Kokan Co., Ltd., 1-1-2
 Marunouchi, Chiyoda-ku, Tokyo

71. Applicant: Nippon Kokan Co., Ltd., 1-1-2 Marunouchi,
 Chiyoda-ku, Tokyo

Japanese → English

Laid-open Patent Application No. Sho 62-278770

Klarquist, Sparkman, LLP

Ref. No. 6454-58227

74.

Agent: Masatoshi Sato, Patent Agent, and two others

SPECIFICATION

1. TITLE OF INVENTION

Method of Recovering CO₂ and H₂ from Converter Gas

2. WHAT IS CLAIMED IS:

A method of recovering CO₂ and H₂ from converter gas in which converter gas is modified, and then the CO₂ in the modified gas is absorbed in an absorption tower and CO₂ is obtained from the absorption solution, and then the absorption tower offgas is pressurized and the pressurized gas is treated by a PSA (pressure swing adsorber), wherein PSA offgas is additionally treated by a PSA installed in series and then supplied to a fuel cell, with electricity obtained and the fuel cell completely combusting the offgas, and the combustion heat is converted to steam and utilized.

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3.

DETAILED DESCRIPTION OF THE INVENTION

[Industrial Field Of Application]

The present invention pertains to a method of recovering CO₂ and H₂ from converter gas generated from an LD converter, etc. when making steel and obtaining heat and electrical energy.

[Prior Art]

In recent years, LD converters, etc. used for making steel have sought to actively recover and effectively utilize the converter exhaust gas, which contains CO, CO₂, and H₂ generated during operations.

These converter gas components, on a volume standard (hereinafter the same) are CO₂ 10~15%, H₂ 1%, CO 65~75%, N₂ 10%, and O₂ 1%.

This converter gas itself can be used as fuel gas, but sometimes a recovery method is performed in which this gas is used as a raw-material gas, with steam added to it to cause a shift reaction that makes carbon dioxide and hydrogen; this increases the added value.

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FIG. 2 is a process diagram of the conventional method of recovering CO₂ and H₂ from LD converter gas.

In the drawing, LD converter gas with the aforesaid composition is used as a raw material and passes through a modification step in which this gas is pressurized to 9~20 kg/cm², steam is added to it, a shift reaction is caused, and the gas components are modified. (More than this is not shown in the drawing.)

Next, the modified gas is introduced to an absorption tower 1, the CO₂ in the modified gas is absorbed in the potassium carbonate solution (K₂CO₃) that is the absorption solution and makes potassium bicarbonate (KHCO₃); this is stripped with steam and the absorption solution becomes K₂CO₃ while the stripped CO₂ gas is made into a product.

The CO₂ is discharged, and the H₂-rich offgas in the absorption tower 1 has its pressure increased from 9~20 kg/cm² to 9~25 kg/cm² by a PSA compressor 2, and is treated by a PSA (Pressure Swing Adsorption device) 3. Gases other than hydrogen are adsorbed in the PSA by utilizing the pressure differential, and separated into product high-

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purity hydrogen (PH_2) and offgas (RH_2). The offgas is stored in an HR_2 holder 4, and is exhausted as fuel for another process by a blower 5.

An example of the components of the offgas in this case are CO_2 12%, H_2 58%, CO 3.5%, N_2 26%, and O_2 0.5%.

[Problems the Invention Is to Solve]

As noted above, even though offgas in the conventional recovery method for recovering CO_2 and H_2 from converter gas contains about 60% H_2 , it is merely to be used for fuel.

Hospitals, offices, etc. in the vicinity of a steel plant need steam, electricity, hot water, heating and cooling facilities, etc. Currently, as described previously, even though the CO_2 , H_2 , etc. recovered from converter gas are a beneficial energy source, there are many cases in which the demand of the aforesaid sort of public welfare and management facilities is met by purchased electricity, and heavy oil, etc. is used for hot water and heating and cooling.

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The object of the present invention is to provide a method of efficiently recovering CO₂ and H₂ from converter gas that is relatively low in calories (500~1700 Kcal/Nm³) for manufacturing equipment but high in hydrogen concentration, and of efficiently utilizing the energy contained therein.

[Means for Solving the Problems]

The present inventors focused on PSA offgas, which contains about 60% hydrogen in the conventional method, studied efficient utilization of this offgas, and arrived at the present invention. That is, the present invention is a method of separating and recovering H₂ from converter gas in which converter gas is modified, and then the CO₂ component in the modified gas is absorbed in K₂CO₃ solution in an absorption tower and product CO₂ is obtained from the absorption solution, and then the absorption waste gas is pressurized and the pressurized gas is treated by a PSA; it is characterized in that PSA offgas is additionally treated by a PSA installed in series and then the gas (a gas with little CO component and a high-concentration H₂ component)

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is supplied to a fuel cell, electricity is obtained, the fuel cell completely combusts the offgas, and the combustion heat is converted to steam and utilized.

[Operation]

In the inventive method of recovering CO₂ and H₂ from converter gas the PSA offgas, which contains about 60% hydrogen, is additionally treated by a PSA installed in series, and it is possible to efficiently recover high-concentration H₂ with a low CO component, so it is possible to supply it as fuel gas for a fuel cell.

Furthermore, the fuel cell completely combusts the offgas and its exhaust heat can be recovered as steam, so it is possible to advantageously recover in a cost-effective manner various types of energy-electricity, steam, fuel gas, etc.-all at once.

Next, embodiments shall be described.

[Embodiments]

FIG. 1 is a process explanatory diagram for inventive method of recovering CO₂ and H₂ from converter gas.

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In FIG. 1, 1~4 are the same as in the aforesaid FIG. 2, so explanation thereof is omitted.

Item 6 is a PSA installed in series, 7 is a vacuum blower, 8 is a blower, 9 is a valve, 10 is a heat exchanger, 11 is an air blower for a combustion, 12 is a fuel cell, 13 is a hydrogen electrode chamber (negative electrode), 14 is an oxygen electrode chamber (positive electrode), 15 is electrolyte, 16 is a pump, 17 is a preheater, 18 is a combustor, 19 is a boiler, and 20 is a chimney.

The inventive method shall be described based on FIG. 1.

The raw-material gas was LD converter gas (H_2 : 0.7%, CO: 69.8%, CO_2 : 14.7%, N_2 : 14.3%) at $1.298 \text{ Nm}^3/\text{H}$. This gas was pressurized to $9\text{--}11 \text{ kg/cm}^2$ by a compressor, and then this gas was modified to hydrogen through a shift reaction using steam, and then the CO_2 in the gas was absorbed in K_2CO_3 solution, the absorption solution was stripped with steam, and product CO_2 was obtained. Next, the absorption tower 1's offgas was pressurized to 16 kg/cm^2 by the PSA

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compressor 2, and treated by the PSA 3.

In the PSA 3, gases other than H₂ were adsorbed by adsorbent in the tower, and [the offgas] was separated into product H₂ and offgas RH₂ containing gases other than H₂.

The offgas RH₂ was temporarily stored in a holder 4, and by controlling a valve 9 was treated at the next PSA 6 by a suitable blower 8 (2000 nm water column).

The treated hydrogen gas from the PSA 6 (553 Nm³/H) was treated by the PSA 6, so it contains H₂ 89.5%, CO 1.1%, CO₂ 0.4%, and N₂ 9%; it is something that can be supplied to a fuel cell.

Next, an embodiment for recovering electricity and steam from the fuel cell shall be described.

The treated hydrogen gas from the aforesaid PSA 6 was sucked in by the vacuum blower 7, and at the heat exchanger 10, the offgas temperature was heat-exchanged with waste gas after recovering combustion exhaust heat from the fuel cell 12's offgas and raised to 90°C, and introduced to the hydrogen electrode (negative electrode) of the fuel cell 12.

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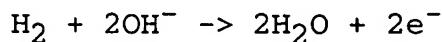
Klarquist, Sparkman, LLP

Ref. No. 6454-58227

Meanwhile, air 2000 nm³/H was supplied to the heat exchanger 10 by the blower 11 and heated to 120°C; this air was introduced to the fuel cell 12's oxygen electrode 14.

The fuel cell 12 uses an aqueous phosphoric acid solution 15 as its electrolyte, and has two electrodes made of platinum and ruthenium surrounding this.

At the negative electrode (hydrogen electrode 13), the introduced oxygen gas passes through tiny holes in the electrode, comes close to the reaction zone, is adsorbed by catalyst added to the electrode, and becomes active atomic hydrogen. This atomic hydrogen reacts with hydroxide ions in the electrolyte as in the following equation and becomes water, and when doing so transports two electrons to the electrode.



At the positive electrode (oxygen electrode 14), the two electrons from the electrode are received, based on the presence of the catalyst, and oxygen molecules in the air supplied from outside react with water from the electrolyte and generate a hydrogen peroxide ion and a hydroxide ion as

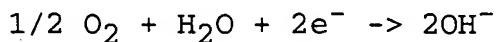
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in the following equation. The hydrogen peroxide ion comes into contact with the catalyst and breaks down and forms a hydroxide ion and oxygen. This oxygen is utilized in the electrode reaction once again. Therefore, the positive electrode reaction is:



The hydroxide ions generated by this positive electrode 14 are transported in the electrolyte 15 and reach the negative electrode 13. This formed an entire circuit and produced 740 KW of electricity.

The 190°C offgasses from both the negative electrode 13 and positive electrode 14 of the fuel cell were introduced to the combustor 18 and completely combusted. The 430°C combustion exhaust gas [was supplied to] the boiler 19, where its waste heat was recovered and 6 kg/cm² of saturated steam 0.7T/H was recovered.

Furthermore, a preheater 17 for pure water was provided inside the negative electrode 13's chamber, and after preheating [this water] was supplied to the boiler 19.

The boiler 19's offgas [was supplied to] the heat exchanger 10 and [used to] preheat the gas supplied to the fuel cell and air as described earlier and then vented from a chimney 20.

As a result, the value of converter gas was increased 1.86 times compared to the conventional method.

[Effect of the Invention]

The following sorts of effects are achieved by the inventive method of recovering CO₂ and H₂ from converter gas.

- (1) Electricity, steam, fuel gas, etc. can be obtained all at once from offgas from converter gas purification equipment, and it is possible to give converter gas higher added value.
- (2) It is possible to respond to a varied demand for electricity, steam, fuel gas, etc., so one can rationalize the facilities in the vicinity of a steel mill.

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4. BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a process explanatory drawing for the inventive method. FIG. 2 is a process explanatory drawing for the conventional method.

In the drawings, 1 is an absorption tower, 2 is a PSA compressor, 3 is a PSA, 4 is a RH₂ holder, 6 is a PSA (offgas treatment), 7 is a vacuum blower, 8 is a blower, 9 is a valve, 10 is a heat exchanger, 11 is an air blower, 12 is a fuel cell, 13 is a negative electrode (hydrogen electrode), 14 is a positive electrode (oxygen electrode), 15 is electrolyte, 17 is a preheater, 18 is a combustor, and 19 is a boiler.

Furthermore, codes that are the same in both drawings indicate equivalent parts.

Agent: Masatoshi Sato, Patent Agent

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FIG. 1

1: Absorption tower
2: PSA compressor
3: PSA
4: RH₂ holder
5: Blower
6: PSA (offgas treatment)
7: Vacuum blower
8: Blower
9: Valve
10: Heat exchanger
11: Air blower
12: Fuel cell
13: Negative electrode (hydrogen electrode)
14: Positive electrode (oxygen electrode)
15: Electrolyte
16: Pump
17: Preheater
18: Combustor
19: Boiler

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20: Chimney

FIG. 2

* * *

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